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## **JAPANESE / ENGLISH TRANSLATION OF**

**Japanese Patent Application JP 9 - 272191 A**

**Multilayer Polyester Sheet, and Container Made Therefrom**

**Your Ref: 102203 - 18**

**For: Eastman Chemical Company**

9109 Eagle Hills Dr . Las Vegas NV 89134 . ph: 866-722-7990 (toll-free) . fax: 702-973-6009 . email: sales@tls-translations.com

(19) Japanese Patent Office (JP)

(11) Unexamined Patent Application No:

**(12) Unexamined Patent Gazette (A)**

**Hei 9-272191**

(43) Date of Publication: October 21, 1997

(51) Int. Cl. <sup>6</sup>	Class. Symbols	Internal Office Registration Nos.:	FI	Technical Classification Field
B32B 27/36 1/02			B32B 27/36 1/02	
//C08K 5/10 7/16	KJV KKF		C08K 5/10 7/16	KJV KKF
C08L 67/02			C08L 67/02	

Request for Examination: Not yet submitted

Number of Claims: 3 FD

(Total of 7 pages [in the original])

(21) Application No.: Hei 8-108545  
(22) Date of Filing: April 3, 1996

(71) Applicant: 000000952  
Kanebo, Ltd.  
17-4, 5-chome Sumida  
Sumida-ku, Tokyo  
(72) Inventor: Tetsuo Kunimaru  
6-10 Unoshima-cho  
Ube-shi, Yamaguchi-ken  
(72) Inventor: Akihide Fujita  
6-6-101 Kanebo-cho  
Bofu-shi, Yamaguchi-ken  
(72) Inventor: Masaki Yamamoto  
5-1-9 Kanebo-cho  
Bofu-shi, Yamaguchi-ken

**(54) [Title of the Invention]**

**Multilayer Polyester Sheet, and Container Made  
Therefrom**

**(57) [Summary]**

**[Object]** To improve the slip properties of a sheet without adversely affecting its transparency by compounding inert particles and an aliphatic ester compound with a polyester to resolve difficulties related to blocking and to cutting the sheet during transport on the production line when manufacturing various types of containers from amorphous polyester (A-PET) sheets.

**[Means of Achievement]** A multilayer polyester sheet wherein a copolyester obtained by compounding specific quantities of inert particles of a specific mean particle diameter (such as silica or calcium carbonate) and a long-chained aliphatic ester compound with polyethylene terephthalate copolymerized with 1,4-cyclohexane dimethanol is used as the skin layer; a

polyester made from a component consisting mainly of ethylene terephthalate units is used as the core layer; and the thickness of the skin layer and the intrinsic viscosity of all layers of the sheet are set within specified ranges; and a polyester container manufactured from this sheet.

#### **[Claims]**

**[Claim 1]** A multilayer polyester sheet comprising (A) a skin layer composed of a copolyester that has an acid component consisting mainly of terephthalic acid, that also has 10-90 mol% ethylene glycol and 90-10 mol% 1,4-cyclohexane dimethanol as the glycol component, and that contains 10-5000 ppm of inert particles with a mean particle diameter of 0.5-30  $\mu\text{m}$  and 0.1-5% by weight of an ester compound having a total of 30-60 carbon atoms and consisting of an aliphatic alcohol and an aliphatic monocarboxylic acid; and (B) a core layer composed of a polyester that has an acid component consisting mainly of terephthalic acid and a glycol component consisting mainly of ethylene glycol, wherein said multilayer polyester sheet is such that the skin layer is laminated on at least one side of the core layer, the total thickness of the skin layer is 3-50% of the combined thickness of the sheet layers, and the intrinsic viscosity of all the layers of the sheet is 0.5-1.1.

**[Claim 2]** The multilayer polyester sheet of Claim 1 wherein the skin layer (A) is composed of a copolyester that has an acid component consisting mainly of terephthalic acid, that also has 10-90 mol% ethylene glycol and 90-10 mol% 1,4-cyclohexane dimethanol as the glycol component, and that contains 10-5000 ppm of inert particles with a mean particle diameter of 0.5-10  $\mu\text{m}$ , 1500 ppm or less of inert particles with a mean particle diameter of 10-30  $\mu\text{m}$ , and 0.1-5% by weight of an ester compound having a total of 30-60 carbon atoms and consisting of an aliphatic alcohol and an aliphatic monocarboxylic acid.

**[Claim 3]** A polyester container obtained by cutting, punching, folding, bonding, and assembling the multilayer polyester sheet of Claim 1 or 2.

#### **[Detailed Description of the Invention]**

**[0001]**

**[Technological Field of the Invention]** The present invention relates to a polyester sheet that can be provided with improved slip properties, cuttability, and solvent bonding properties without any adverse effect on the transparency.

[0002]

[Prior Art] Saturated polyesters, especially crystalline polyester resins represented by polyethylene terephthalate (abbreviated hereinafter as PET), are widely used as polymers for fibers, sheets, and films. They have also come to be used in beverage bottles for carbonated beverages, juices, and beer; cosmetics containers; and food trays due to their excellent chemical resistance and low gas permeability.

[0003] Among these, amorphous polyester sheets referred to as "A-PET" have drawn attention for their excellent recyclability, low pollution, and food safety, and are being used in rapidly increasing quantities as substitute packaging materials for vinyl chloride and polystyrene. In addition to being used as thermoformed containers for foods and drugs and as blister packs for miscellaneous goods, these polyester sheets are being employed as clear cases for cosmetics and electrical equipment due to their excellent transparency.

[0004] Using such polyester sheets as clear cases in the past caused blocking to occur between the sheets during transport on the production line due to the poor slip properties of the sheets, difficulties to be experienced when cutting the sheets, and other problems to be encountered, and improvement was therefore desired.

[0005] Many methods have been proposed for improving the slip properties of polyester sheets. For example, methods such as those described in JP (Kokai) 53-14753 and 4-136263 are known as methods of precipitating internal particles and adding inert particles. However, it is difficult to regard these methods as satisfactory because adequate slip properties cannot be obtained if the amount added is not increased and because the transparency declines if the amount added becomes too high.

[0006] There are also methods that involve applying silicone to the surface. However, such methods cannot be regarded as satisfactory since it is difficult to accomplish uniform application and drying with this method, the properties of the product are not stable, and the printability and adhesiveness are reduced.

[0007] A sheet that uses high-molecular-weight linear polyester obtained by copolymerizing a bivalent alicyclic alcohol together with an aromatic dicarboxylic acid and a bivalent aliphatic alcohol in the surface layer and backing layer has also been proposed (JP (Kokai) 5-162271). This method is aimed at resolving the aforementioned problems by affixing a PET sheet that

contains special components as the surface layer. However, the slip properties of the surface cannot be regarded as adequate with such as method.

**[0008]**

**[Problems to Be Solved by the Invention]** As a result of studies conducted to resolve such past problems, the present inventors discovered that excellent slip properties, scratch resistance, cuttability, and adhesiveness can be imparted without any adverse effect on the inherent transparency of A-PET by compounding a specific quantity of inert particles with the polyester, and also compounding a specific ester compound composed of an aliphatic alcohol and an aliphatic monocarboxylic acid.

**[0009]**

**[Means Used to Solve the Above-Mentioned Problems]** Specifically, the present invention is, first, a multilayer polyester sheet comprising (A) a skin layer composed of a copolyester that has an acid component consisting mainly of terephthalic acid, that also has 10-90 mol% ethylene glycol and 90-10 mol% 1,4-cyclohexane dimethanol as the glycol component, and that contains 10-5000 ppm of inert particles with a mean particle diameter of 0.5-30  $\mu\text{m}$  and 0.1-5% by weight of an ester compound having a total of 30-60 carbon atoms and consisting of an aliphatic alcohol and an aliphatic monocarboxylic acid; and (B) a core layer composed of a polyester that has an acid component consisting mainly of terephthalic acid and a glycol component consisting mainly of ethylene glycol, wherein the multilayer polyester sheet is such that the skin layer is laminated on at least one side of the core layer, the total thickness of the skin layer is 3-50% of the combined thickness of the sheet layers, and the intrinsic viscosity of all layers of the sheet is 0.5-1.1.

**[0010]** The second invention is the aforementioned first multilayer polyester sheet in which the skin layer (A) is composed of a copolyester that has an acid component consisting mainly of terephthalic acid, that also has 10-90 mol% ethylene glycol and 90-10 mol% 1,4-cyclohexane dimethanol as the glycol component, and that contains 10-5,000 ppm of inert particles with a mean particle diameter of 0.5-10  $\mu\text{m}$ , 1500 ppm or less of inert particles with a mean particle diameter of 10-30  $\mu\text{m}$ , and 0.1-5% by weight of an ester compound having a total of 30-60 carbon atoms and consisting of an aliphatic alcohol and an aliphatic monocarboxylic acid.

[0011] The third invention is a polyester container obtained by cutting, punching, folding, bonding, and assembling the multilayer polyester sheet of the aforementioned first or second invention.

### **[Embodiments of the Invention]**

[0012] Examples of the polyester used as the core layer (B) of the present invention include homo-PET as well as copolyesters in which part of the terephthalic acid component has been substituted by one or more types of dicarboxylic acid component, such as isophthalic acid, adipic acid, diphenyl carboxylic acid, diphenyl ether dicarboxylic acid, sebacic acid, or naphthalene dicarboxylic acid, and/or part of the ethylene glycol component has been substituted by one or more types of glycol component, such as diethylene glycol, hexamethylene glycol, trimethylene glycol, propylene glycol, cyclohexane dimethanol, neopentyl glycol, or butylene glycol.

[0013] The polyester may also contain a trifunctional or higher compound, or a monofunctional compound as a polymer component within a range in which substantial linearity is believed to exist. Heat stabilizers, fluidity improving agents, ultraviolet absorbers, antistatic agents, anticondensation agents, and the like can also be added to the polyester as long as its transparency is not compromised. Colorants such as titanium dioxide, calcium carbonate, iron oxide, and carbon black can also be contained when delustering is desired.

[0014] The thickness of the sheet used in the present invention is not particularly restricted, but is usually 50-1500  $\mu\text{m}$ , and is preferably 100-500  $\mu\text{m}$ .

[0015] The intrinsic viscosity of the multilayer polyester sheet of the present invention must be 0.5-1.1. The inherent impact strength of the polyester sheet drops when the viscosity is less than 0.5. Not only does the economic aspect deteriorate when the viscosity exceeds 1.1, but melt extrusion becomes difficult during sheet manufacture.

[0016] The copolyester used as skin layer (A) in the present invention has an acid component consisting mainly of terephthalic acid, and is composed of a glycol component that comprises 10-90 mol% of ethylene glycol and 90-10 mol% of 1,4-CHDM. 1,4-CHDM is needed as the copolymer component used to make it possible to impart excellent adhesiveness and excellent impact resistance, particularly impact resistance over time. Not only is the adhesiveness inferior when the 1,4-CHDM content is less than 10 mol% or more than 90 mol%, but the compatibility with the ester compound used in the present invention decreases and the transparency is adversely affected. Various dicarboxylic acid components and/or various glycol components

may also be copolymerized within the ranges that do not compromise the properties of the polymer.

[0017] The term "inert particles used in the present invention" refers to microparticles that remain inert in the polyester synthesis reaction system. Specifically, these are microparticles such as silica, calcium carbonate, aluminum silicate, barium sulfate, calcium phosphate, talc, titanium dioxide, aluminum hydroxide, calcium terephthalate, calcium silicate, fluorine-containing microparticulate polymers, and crosslinked polystyrene polymers.

[0018] These inert particles may be used individually or in combinations of two or more types. Methods employed to uniformly disperse the particles throughout a sheet include those in which the particles are added as a slurry during polymerization, and those in which a master batch is first prepared by melt kneading the inert particles and resin, and this master batch is then fed to produce a prescribed amount of inert particles during sheet manufacture. The inert particles may also be surface treated with olefin wax, aliphatic acid, aliphatic acid metal salt, and the like within the range that does not lower the transparency of the sheet.

[0019] The present invention is characterized by adding 10-5000 ppm of inert particles with a mean particle diameter of 0.5-30  $\mu\text{m}$  to the resin (A) used in the skin layer. The slip properties become unsatisfactory when the content of inert particles is less than 10 ppm. The transparency is adversely affected when there is more than 5,000 ppm. If even better slip properties and transparency are required, it is preferable to use 10-5,000 ppm of particles with a mean particle diameter of 0.5-10  $\mu\text{m}$ , and 1500 ppm or less of inert particles with a mean particle diameter of 10-30  $\mu\text{m}$ . Joint use of two types of inert particles in this way yields a sheet with even better slip properties and scratch resistance than when only one type of inert particle is used alone.

[0020] A quantity of 0.1-5% by weight of an ester compound having a total of 30-60 carbon atoms and consisting of an aliphatic alcohol and an aliphatic monocarboxylic acid must be used in the present invention. The aliphatic alcohol that constitutes the aforementioned ester compound preferably has 6 or more carbon atoms. Examples include, but are not limited to, lauryl alcohol, myristyl alcohol, cetyl alcohol, heptadecyl alcohol, stearyl alcohol, nonadecyl alcohol, eicosyl alcohol, behenyl alcohol, ceryl alcohol, and melissyl alcohol.

[0021] The aliphatic monocarboxylic acid that constitutes the aforementioned ester compound preferably has 10 or more carbon atoms. Examples include, but are not limited to, myristic acid,



palmitic acid, stearic acid, arachidonic acid, behenic acid, lignoceric acid, cerotic acid, montanic acid, and melissic acid.

[0022] The ester compound with a total of 30-60 carbon atoms composed of the aliphatic alcohol and aliphatic monocarboxylic acid used in the present invention is produced by appropriately selecting and compounding the aforementioned aliphatic alcohol and aliphatic monocarboxylic acid. Specific examples include lauryl stearate, stearyl stearate, lauryl melissate, behenyl behenate, and ceryl melissate. The heat resistance becomes inadequate, decomposition occurs, and film formation becomes difficult during the manufacture of the multilayer polyester sheet when the ester compound has fewer than 30 carbon atoms. Compatibility with the copolyester declines and the transparency is adversely affected when the compound has more than 60. There is little bleed-out to the surface of the sheet and satisfactory slip properties cannot be obtained when the compounded amount is less than 0.1% by weight. Adhesiveness deteriorates due to excessive bleed-out to the surface of the sheet, and the transparency also decreases when the compounded amount exceeds 5% by weight.

[0023] Known methods can be used without any particular restrictions for adding the above ester compound. It is possible, for example, to prepare a master batch, to supply it during the process of extrusion into pellets, and to add the compound during polymerization.

[0024] The layer ratio of the multilayer polyester sheet of the present invention must be such that the thickness of the entire skin layer is 3-50% of the combined thickness of the sheet layers. It becomes difficult to form a uniform skin layer during sheet manufacture when the thickness is less than 3%. Not only is a ratio greater than 50% not economical, but also the transparency is adversely affected.

[0025] The multilayer polyester sheet of the present invention can be manufactured by known technology such as melt lamination, coextrusion, and dry lamination. However, coextrusion is preferred in terms of the quality of the sheet. A coextruded multilayer sheet can be obtained, for example, by a method in which the material is melt-extruded using an ordinary polyester extruder such as a twin-screw vented extruder, and the molten resin is cooled on a cooling drum. Cooling the sheet as rapidly as possible is preferred to prevent a decline in transparency due to crystallization. The degree of crystallization is preferably 10% by weight or lower (density 1.348 g/cm<sup>3</sup> or higher), and the sheet haze is preferably 5% or lower.

[0026] Methods of film formation include those in which the material is sandwiched and cooled between metal rolls (touch-roll method), those in which static electricity is applied, and those in which an air knife is used. However, the touch-roll method is preferred from the viewpoint of sheet luster and thickness uniformity.

[0027] In-line recycling by crushing and reusing as raw materials the scraps produced when cutting the sheet to the prescribed width, and the skeletons and other types of waste produced when punching out products is commonly employed for A-PET. Such techniques can also be used in the present invention within the range that does not greatly lower the intrinsic viscosity of the sheet.

[0028] The sheet obtained in this way can be made into a clear case by being cut, punched, folded, bonding, and assembled in accordance with the usual methods.

[0029]

[**Merits of the Invention**] The present invention improves the slip properties when cutting the sheet and improves the blocking of adjacent sheets and the scratch resistance of the sheets while maintaining the transparency of A-PET when the sheets are made into a product. The invention is also suitable for improving the blocking resistance of a polyester container made by cutting, punching, folding, bonding, and assembling such sheets, and for improving the blocking resistance of a thermoformed product.

[0030]

[**Working Examples**] The present invention is explained more concretely below through working examples. However, the present invention is not limited to these. The characteristic values were measured by the following methods.

[0031] (1) Intrinsic viscosity

Measured at a concentration of 1.0 g/dL at 20°C using a phenol/tetrachloroethane mixed solvent with a weight ratio of 60/40.

[0032] (2) Sheet haze

The haze of the sheet was measured according to the method of JIS-K-7105 using a haze meter NDH-20D.

[0033] (3) Coefficient of static friction

The coefficient of static friction of the sheet was measured according to the method of ASTM-1894, using a friction tester TR made by Toyo Seiki.

[0034] (4) Cuttability

Evaluated by stacking 50 sheets 200 mm long × 200 mm wide and cutting the stacks with a Thomson blade.

No. of sheets cut	Ranking
41-50	○
21-40	△
0-20	×

After having been measured for the coefficient of static friction, the sample was measured according to the method of JIS-K-7105 using a haze meter NDH-20D, and the haze of the sheet was evaluated.

Haze	Ranking
Less than 3.5%	○
3.5% or greater	×

[0036] (6) Solvent bonding properties

The sheet was bonded using cyclohexanone, and the peel strength was measured according to the method of JIS-K-6772.

Peel strength	Ranking
>2000 g/15 mm	○
2000-500 g/15 mm	△
Less than 500 g/15 mm	×

**[0037] (7) Mean particle diameter**

The diameter (particle diameter) of the integrated 50% volume in an equivalent sphere distribution measured by a Multisizer (made by Coulter Electronics Ltd.) was taken as the mean particle diameter.

**[0038] Working Examples 1-6 and Comparative Examples 1-13**

Poly-1,4-cyclohexane dimethylene terephthalate resin with an intrinsic viscosity of 0.70 was dried to a water content of 200 ppm or less and melt kneaded by means of a twin-screw mixer with inert particles having the diameters shown in Table 1 at 240°C, yielding a 10-wt% master batch. 10-wt% master batches of the ester compounds of aliphatic alcohols and aliphatic monocarboxylic acids shown in Table 1 were prepared in the same way. Next, multilayer melt extrusion was performed by means of a T-die at 285°C using a twin-screw vented extruder while the degree of vacuum in the vented portion was kept at 5 mmHg. The master batches of these inert particles and esters were compounded in prescribed quantities with the copolyester resins in Table 1 during film formation by the touch-roll method, and three-layered, 250-μm-thick sheets with the compositions shown in Tables 1 and 2 were produced. Table 3 shows the properties of these sheets.

**[0039]**

**[Table 1]**

Working example	Sheet properties			Composition of additives and particles					
	Skin layer	Core layer	Skin layer thickness (vs. all layers) (%)	Inert particles (1)		Inert particles (2)		Additives	
	Type	Type		Type Mean particle diameter	Amount compounded (ppm)	Type Mean particle diameter	Amount compounded (ppm)	Type	Amt. compounded wt%
1	1,4-CHDM 33 mol% copolymerized	Homo-PET	10	PS cross-linked particles 8 μm	800	Silica gel 20 μm	500	Behenyl behenate	0.6
2	1,4-CHDM 12 mol% copolymerized	Homo-PET	12	PS cross-linked particles 3 μm	1500	Silica gel 20 μm	300	Stearyl stearate	0.8
3	1,4-CHDM 30 mol% copolymerized	Homo-PET	20	PS cross-linked particles 8 μm	2000	—	—	Behenyl behenate	0.7
4	1,4-CHDM 30 mol% copolymerized	Isophthalic acid 5 mol% copolymerized	40	PS cross-linked particles 6 μm	1000	Silica gel 15 μm	1000	Stearyl stearate	0.6
5	1,4-CHDM 12 mol% copolymerized	Homo-PET	20	Silica gel 8 μm	2500	PS cross-linked particles 20 μm	200	Melissyl melissate	0.4
6	1,4-CHDM 30 mol% copolymerized	Homo-PET	20	PS cross-linked particles 8 μm	3000	Silica gel 20 μm	300	Behenyl behenate	3.0

1,4-CHDM: 1,4-cyclohexane dimethanol  
PS: Polystyrene

[0040]

[Table 2]

Comparative example	Sheet properties			Composition of additives and particles					
	Skin layer	Core layer	Skin layer thickness (vs. all layers) (%)	Inert particles (1)		Inert particles (2)		Additives	
	Type	Type		Type Mean particle diameter	Amount compounded (ppm)	Type Mean particle diameter	Amt. com-bined (ppm)	Type	Amt. compounded wt%
1	—	Homo-PET	—	—	—	—	—	—	—
2	1,4-CHDM 30 mol% copolymerized	Homo-PET	12	—	—	—	—	Behenyl behenate	0.7
3	1,4-CHDM 30 mol% copolymerized	Homo-PET	10	—	—	Silica gel 20 μm	7500	Behenyl behenate	0.6
4	1,4-CHDM 30 mol% copolymerized	Isophthalic acid 5 mol% copolymerized	10	PS cross-linked particles 8 μm	2000	Silica gel 20 μm	300	—	—
5	1,4-CHDM 12 mol% copolymerized	Homo-PET	10	PS cross-linked particles 3 μm	1500	Silica gel 50 μm	300	Stearyl stearate	0.6
6	1,4-CHDM 12 mol% copolymerized	Homo-PET	10	PS cross-linked particles 8 μm	1500	Silica gel 20 μm	300	Stearyl stearate	9.0
7	1,4-CHDM 30 mol% copolymerized	Homo-PET	20	Silica gel 0.05 μm	2500	—	—	Stearyl stearate	0.8
8	1,4-CHDM 30 mol% copolymerized	Homo-PET	20	PS cross-linked particles 8 μm	1500	Silica gel 20 μm	300	Ethyl laurate	0.8
9	1,4-CHDM 30 mol% copolymerized	Homo-PET	12	PS cross-linked particles 6 μm	1500	Silica gel 20 μm	300	Melissyl hentariacon-tanate [phonetic]	0.7
10	1,4-CHDM 95 mol% copolymerized	Homo-PET	12	PS cross-linked particles 8 μm	1500	PS cross-linked particles 20 μm	500	Behenyl behenate	0.7
11	1,4-CHDM 3 mol% copolymerized	Homo-PET	12	PS cross-linked particles 6 μm	2500	Silica gel 20 μm	500	Behenyl behenate	1.5
12	1,4-CHDM 30 mol% copolymerized	Homo-PET	2	PS cross-linked particles 8 μm	2500	PS cross-linked particles 20 μm	500	Behenyl behenate	0.05
13	1,4-CHDM 30 mol% copolymerized	Homo-PET	60	PS cross-linked particles 8 μm	2500	Silica gel 20 μm	500	Behenyl behenate	0.7

1,4-CHDM: 1,4-cyclohexane dimethanol

PS: polystyrene

[0041]

[Table 3]

	IV	Haze (%)	Friction coefficient ( $\mu$ )	Cuttability	Scratch resistance	Solvent bonding properties
Working example 1	0.65	1.5	0.36	○	○	○
Working example 2	0.70	1.6	0.34	○	○	○
Working example 3	0.70	1.6	0.36	○	○	○
Working example 4	0.72	2.0	0.32	○	○	○
Working example 5	0.62	1.8	0.36	○	○	○
Working example 6	0.75	2.6	0.30	○	○	○
Comparative example 1	0.67	0.6	>1	×	○	×
Comparative example 2	0.65	1.1	0.46	×	○	○
Comparative example 3	0.65	6.0	0.33	○	×	○
Comparative example 4	0.70	1.1	0.68	×	○	○
Comparative example 5	0.70	2.0	0.36	○	×	○
Comparative example 6	0.70	10.0	0.28	○	○	×
Comparative example 7	0.65	2.5	0.42	×	○	○
Comparative example 8	0.65	1.0	0.62	×	○	○
Comparative example 9	0.45	4.8	0.38	×	○	○
Comparative example 10	0.55	5.0	0.35	○	○	×
Comparative example 11	0.70	4.6	0.36	○	○	×
Comparative example 12	0.70	1.2	0.65	×	○	○
Comparative example 13	0.70	3.5	0.34	○	○	○